

WHAT IS CLAIMED IS:

1. A method of producing a laminated dielectric element by alternately laminating dielectric ceramic layers containing lead in the composition thereof and base metal electrode layers of a base metal, comprising the steps of:

5 printing electrodes by applying a paste material for the base metal electrodes containing an oxide of a base metal onto the surfaces of at least one 10 side of the ceramic green sheets obtained by molding, into the form of a sheet, a ceramic material of a metal oxide containing a lead oxide;

15 laminating and press-adhering the ceramic green sheets onto which the paste material for the base metal electrodes is applied to fabricate a laminate thereof;

20 reducing the electrodes to form base metal electrode layers by heating the laminate in a heating furnace while flowing an atmospheric gas and by reducing the paste material for the base metal electrodes; and

sintering the laminate;

25 wherein in the step of reducing the electrodes, the paste material for the base metal electrodes is so reduced that the amount of the base metal oxide remaining in the base metal electrode layers is not larger than 20% by weight and that the amount of lead liberated from the ceramic material is not larger than 30 atomic % within 5000 Å from the surfaces of the base metal electrode layers.

30 2. A method of producing a laminated dielectric element according to claim 1, wherein the atmospheric gas in the step of reducing the electrodes contains a reducing gas for reducing the paste material for the base metal electrodes and an oxidizing gas of oxygen and/or a 35 gas that forms oxygen.

3. A method of producing a laminated dielectric element according to claim 2, wherein the reducing gas is

hydrogen and the oxidizing gas is oxygen.

4. A method of producing a laminated dielectric element according to claim 3, wherein, in the step of reducing the electrodes, the heating starts at a 5 temperature-raising rate of not larger than 200°C/h, a maximum holding temperature is maintained at 300°C to 400°C for 0.5 to 15 hours, the furnace is cooled, and the laminate is taken out from the heating furnace at a 10 temperature of not higher than 90°C while maintaining the oxygen partial pressure in the atmospheric gas to be from  $1 \times 10^{-23.9}$  to  $1 \times 10^{-22}$  atm from the start of heating until the laminate is taken out from the heating furnace.

5. A method of producing a laminated dielectric element according to claim 3, wherein if the total amount 15 of flow rates of gases constituting the atmospheric gas is regarded to be the total flow rate  $F$  (ml/min), then, an oxygen flow rate  $f_o$  (ml/min) and a hydrogen flow rate  $f_H$  (ml/min) in the step of reducing the electrodes have a relationship of oxygen flow rate  $f_o = A \times$  hydrogen flow 20 rate  $f_H$ /total flow rate  $F - B$  (where  $20 \leq A \leq 24$ ,  $12 \leq B \leq 16$ ,  $8 \leq A - B \leq 8.22$ ).

6. A method of producing a laminated dielectric element according to claim 5, wherein, in holding the maximum temperature in the step of reducing the 25 electrodes, if the molar flow rate of water formed by the reaction of hydrogen with oxygen is denoted by  $W$  and the molar flow rate of excess of hydrogen that is remaining by  $H$ , the values of the two molar flow rates lie within the following ranges,

30  $0.0012 \leq H \leq 0.0018$  ( $H$ : mols/min)  
 $0.0002 \leq W \leq 0.001$  ( $W$ : mols/min).

7. A method of producing a laminated dielectric element according to claim 6, wherein, in holding the maximum temperature in the step of reducing the 35 electrodes, an integrated value  $h$  of the molar flow rate of hydrogen during the holding time lies within the

following range,

29000  $\leq$  h  $\leq$  31000 (h: mols/min).

8. A method of producing a laminated dielectric element according to claim 1, wherein, in the step of printing electrodes, the paste for the base metal electrodes is applied maintaining a thickness of 2 to 14  $\mu$ m.

9. A method of producing a laminated dielectric element according to claim 1, wherein the step of reducing the electrodes and the step of sintering are simultaneously conducted by one heating.

10. A method of producing a laminated dielectric element according to claim 1, wherein the base metal is copper.

15. A method of producing a laminated dielectric element according to claim 1, wherein after the step of press-adhesion, the step of dewaxing is conducted to remove organic materials contained in the laminate.

20. A method of producing a laminated dielectric element by alternately laminating dielectric ceramic layers containing lead in the composition thereof and base metal electrode layers of a base metal, comprising the steps of:

25. printing electrodes by applying a paste material for the base metal electrodes containing an oxide of a base metal onto the surfaces of at least one side of ceramic green sheets obtained by molding, into the form of a sheet, a ceramic material of a metal oxide containing a lead oxide;

30. laminating and press-adhering the ceramic green sheets onto which the paste material for the base metal electrodes is applied to fabricate a laminate thereof;

35. reducing the electrodes to form base metal electrode layers by heating the laminate in a heating furnace while flowing an atmospheric gas and by reducing

the paste material for the base metal electrodes; and  
sintering the laminate;  
wherein in the step of reducing the  
electrodes, if the amount of the base metal oxide  
5 remaining in the base metal electrode layers is denoted  
by M (% by weight) (where the remaining amount M is  
determined by a thin-film X-ray diffraction method) and  
if the amount of lead liberated from the ceramic material  
within 5000 Å from the surfaces of the base metal  
10 electrode layers is denoted by N (atomic %) (where the  
amount N of lead is determined by an X-ray photoelectron  
spectroscopic method within a range of 5000 Å from the  
surfaces of the base metal electrode layers), then, there  
is the relationship,

15 
$$N = C \times M + D, \quad 0 \leq M < 20$$

where C and D are values independent from  
M and N, and

lie within ranges of  $-1.5 \leq C < -1.0$  and  
 $30 \leq D < 36$ ,

20 respectively,

between M (% by weight) and N (atomic %).

13. A method of producing a laminated dielectric  
element according to claim 12, wherein the atmospheric  
gas in the step of reducing the electrodes contains a  
25 reducing gas for reducing the paste material for the base  
metal electrodes and an oxidizing gas of oxygen and/or a  
gas that forms oxygen.

14. A method of producing a laminated dielectric  
element according to claim 13, wherein the reducing gas  
30 is hydrogen and the oxidizing gas is oxygen.

15. A method of producing a laminated dielectric  
element according to claim 14, wherein, in the step of  
reducing the electrodes, the heating starts at a  
temperature-raising rate of not larger than 200°C/h, a  
35 maximum holding temperature is maintained at 300°C to  
400°C for 0.5 to 15 hours, the furnace is cooled, and the

5       laminate is taken out from the heating furnace at a temperature of not higher than 90°C while maintaining the oxygen partial pressure in the atmospheric gas to be from  $1 \times 10^{-23.9}$  to  $1 \times 10^{-22}$  atm from the start of heating until the laminate is taken out from the heating furnace.

10      16. A method of producing a laminated dielectric element according to claim 14, wherein if the total amount of flow rates of gases constituting the atmospheric gas is regarded to be the total flow rate  $F$  (ml/min), then, an oxygen flow rate  $f_o$  (ml/min) and a hydrogen flow rate  $f_H$  (ml/min) in the step of reducing the electrodes have a relationship of oxygen flow rate  $f_o = A \times$  hydrogen flow rate  $f_H$ /total flow rate  $F - B$  (where  $20 \leq A \leq 24$ ,  $12 \leq B \leq 16$ ,  $8 \leq A - B \leq 8.22$ ).

15      17. A method of producing a laminated dielectric element according to claim 16, wherein, in holding the maximum temperature in the step of reducing the electrodes, if the molar flow rate of water formed by the reaction of hydrogen with oxygen is denoted by  $W$  and the molar flow rate of excess of hydrogen that is remaining by  $H$ , the values of the two molar flow rates lie within 20 the following ranges,

0.0012  $\leq H \leq 0.0018$  (H: mols/min)

0.0002  $\leq W \leq 0.001$  (W: mols/min).

25      18. A method of producing a laminated dielectric element according to claim 17, wherein, in holding the maximum temperature in the step of reducing the electrodes, an integrated value  $h$  of the molar flow rate of hydrogen during the holding time lies within the 30 following range,

29000  $\leq h \leq 31000$  (h: mols/min).

35      19. A method of producing a laminated dielectric element according to claim 12, wherein, in the step of printing electrodes, the paste for the base metal electrodes is applied maintaining a thickness of 2 to 14  $\mu\text{m}$ .

20. A method of producing a laminated dielectric element according to claim 12, wherein the step of reducing the electrodes and the step of sintering are simultaneously conducted by one heating.

5 21. A method of producing a laminated dielectric element according to claim 12, wherein the base metal is copper.

10 22. A method of producing a laminated dielectric element according to claim 12, wherein after the step of press-adhesion, the step of dewaxing is conducted to remove organic materials contained in the laminate.

15 23. A method of producing a laminated dielectric element by alternately laminating dielectric ceramic layers and base metal electrode layers of a base metal, comprising the steps of:

20 printing electrodes by applying a paste material for the base metal electrodes containing an oxide of a base metal onto the surfaces of at least one side of ceramic green sheets obtained by molding, into the form of a sheet, a ceramic material;

laminating and press-adhering the ceramic green sheets onto which the paste material for the base metal electrodes is applied to fabricate a laminate thereof;

25 reducing the electrodes to form base metal electrode layers by heating the laminate while flowing an atmospheric gas and by reducing the paste material for the base metal electrodes; and

sintering the laminate;

30 wherein in the step of reducing the electrodes, the atmospheric gas contains a reducing gas for reducing the paste material for the base metal electrodes and an oxidizing gas of oxygen and/or a gas that forms oxygen; and

35 in the step of reducing the electrodes, the heating is conducted with the temperature near a eutectic point of the base metal in the paste material

for the base metal electrodes and of at least one metal in the ceramic green sheets as a maximum holding temperature, and the reducing gas at the maximum holding temperature is contained in an amount of not smaller than 5 0.2% by volume but not larger than 3% by volume in the atmospheric gas.

24. A method of producing a laminated dielectric element according to claim 23, wherein the step of reducing the electrodes and the step of sintering are 10 simultaneously conducted by one heating.

25. A method of producing a laminated dielectric element according to claim 23, wherein the atmospheric gas further contains an inert gas, and the total amount 15 of the reducing gas, the oxidizing gas and the inert gas is not smaller than 99% by volume of the atmospheric gas.

26. A method of producing a laminated dielectric element according to claim 23, wherein the reducing gas is hydrogen and the oxidizing gas is oxygen.

27. A method of producing a laminated dielectric element according to claim 23, wherein if a eutectic point of the base metal in the paste material for the base metal electrodes and of at least one metal in the ceramic green sheets is denoted by  $tm$  ( $^{\circ}C$ ), a maximum holding temperature  $T$  ( $^{\circ}C$ ) in the step of reducing the electrodes has a relationship  $tm - 16 < T < tm + 14$ . 20 25

28. A method of producing a laminated dielectric element according to claim 23, wherein the ceramic material contains lead as an element component.

29. A method of producing a laminated dielectric element according to claim 23, wherein the base metal is 30 copper.

30. A method of producing a laminated dielectric element according to claim 23, wherein after the step of press-adhesion, the step of dewaxing is conducted to 35 remove organic materials contained in the laminate.

31. A method of producing a laminated dielectric element by alternately laminating dielectric ceramic

layers and base metal electrode layers of a base metal, comprising the steps of:

5 printing electrodes by applying a paste material for the base metal electrodes containing an oxide of a base metal onto the surfaces of at least one side of ceramic green sheets obtained by molding, into the form of a sheet, a ceramic material;

10 laminating and press-adhering the ceramic green sheets onto which the paste material for the base metal electrodes is applied to fabricate a laminate thereof;

15 reducing the electrodes to form base metal electrode layers by heating the laminate while flowing an atmospheric gas and by reducing the paste material for the base metal electrodes; and

sintering the laminate;

20 wherein in the step of reducing the electrodes, the atmospheric gas contains a reducing gas for reducing the paste material for the base metal electrodes and an oxidizing gas of oxygen and/or a gas that forms oxygen; and

25 in the step of reducing the electrodes, the heating is conducted with the temperature near a eutectic point of the base metal in the paste material for the base metal electrodes and of at least one metal in the ceramic green sheets as a maximum holding temperature, and the reducing gas in the atmospheric gas at the maximum holding temperature is permitted to flow at a linear density of not smaller than 0.0376 cm/min, 30 but not larger than 0.564 cm/min.

32. A method of producing a laminated dielectric element according to claim 31, wherein the step of reducing the electrodes and the step of sintering are simultaneously conducted by one heating.

35 33. A method of producing a laminated dielectric element according to claim 31, wherein the atmospheric gas further contains an inert gas, and the total amount

of the reducing gas, the oxidizing gas and the inert gas is not smaller than 99% by volume of the atmospheric gas.

34. A method of producing a laminated dielectric element according to claim 31, wherein the reducing gas is hydrogen and the oxidizing gas is oxygen.

35. A method of producing a laminated dielectric element according to claim 31, wherein if a eutectic point of the base metal in the paste material for the base metal electrodes and of at least one metal in the ceramic green sheets is denoted by  $tm$  ( $^{\circ}C$ ), a maximum holding temperature  $T$  ( $^{\circ}C$ ) in the step of reducing the electrodes has a relationship  $tm - 16 < T < tm + 14$ .

36. A method of producing a laminated dielectric element according to claim 31, wherein the ceramic material contains lead as an element component.

37. A method of producing a laminated dielectric element according to claim 31, wherein the base metal is copper.

38. A method of producing a laminated dielectric element according to claim 31, wherein after the step of press-adhesion, the step of dewaxing is conducted to remove organic materials contained in the laminate.

39. A laminated dielectric element fabricated by a production method of claim 1.

40. A laminated dielectric element fabricated by a production method of claim 12.

41. A laminated dielectric element fabricated by a production method of claim 23.

42. A laminated dielectric element fabricated by a production method of claim 31.